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Overview of bromodiphenylether data in aquatic biota and sediments

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OVERVIEW OF BROMODIPHENYLETHER DATA IN AQUATIC BIOTA AND SEDIMENTS

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Figure 1: Sample locations at the North Sea and the north-west Atlantic.

Figure 2: Sample locations in the Netherlands.

Abstract

Concentrations of 2,4,2',4'-tetrabromodiphenylether (TBDE) and 2,4,5,2',4'-pentabromodiphenylether (PeBDE) have been determined in aquatic biota and sediments from 106 different locations in the Netherlands and the north-west Atlantic. 95% Confidence intervals of the data, obtained after analysis of pooled samples, vary between 20% and 50%, dependent of the type of component and the organism.
Most TBDE-concentrations in fish and shellfish are lower than 100 µg/kg. Extremely high levels found in organs of a cormorant suggest a strong biomaginification of PBDEs in these birds. Also for marine mammals biomaginification is observed.
Temporal trends are generally decreasing, although in the rivers Rhine and Meuse a stabilisation is observed since 1988/1989. An increasing trend is found for TBDE-concentrations in eel from the river Roer.
In sediments higher PeBDE-concentrations are found. In comparison with PCBs, for PBDEs a better absorption to sediments is found.

1. Introduction

Polybrominated diphenylethers (PBDEs) are widely used as flame retardants in textiles, paints and plastics in Japan, Western Europe and the USA (Watanabe et al., 1987) In the Netherlands the annual consumption is estimated at 2,500 metric tonnes (Klingenberg, 1989). In Japan the PBDE consumption increased from 2,500 to 22,100 metric tons per year between 1975 and 1987 (Watanabe and Tatsukawa, 1989). In Sweden the PBDE consumption varies between 1,400 and 2,200 metric ton per year (Svenson and Hellsten, 1989).
Because of the wide production and application of PBDEs, its toxicity and environmental persistency (Pijnenburg et al., 1992), PBDEs are considered to be a potential threat for human health, especially through fish consumption. Therefore, the Netherlands Institute for Fisheries Research (RIVO-DLO) initiated analyses of PBDEs in fish and shellfish from the Netherlands and from marine areas that are important grounds for Dutch consumption fish. PBDEs were regularly analysed in yellow eel, in a monitoring programme on concentrations of halogenated organic contaminants in yellow eel from inland waters of the Netherlands (de Boer and Hagel, 1993), and in cod liver from the North Sea, in a monitoring programme on concentrations of halogenated organic contaminants in cod liver from the North Sea (de Boer, 1988a, 1989).
Next to these programmes, a variety of other aquatic organisms and some sediment samples have been analysed on PBDEs. In total samples from 106 different locations have been analysed. This report gives an overview of all data obtained. Temporal trends graphs have been constructed for some locations from the two monitoring programmes.

2. Materials and methods

2.1. Samples

Most samples were taken by staff of the institute. Freshwater fishes were sampled by electrofishing. Sampling of yellow eel was carried out between April and July in order to obtain a sample representative for the location (de Boer and Hagel, 1993). Most marine fishes were sampled by the FRV TRIDENS. Some freshwater and marine fish samples were obtained from local fishermen or from the fishmarket. The 1990 dolphin and harbour porpoise samples were obtained from Dr. R. Kastelein of the Marine Mammal Park, Harderwijk, the Netherlands, and the cormorant sample was obtained from the Central Institute for Veterinary Science, Lelystad, the Netherlands.
Only for yellow eel attention was paid to select a certain length class: in order to restrict the variation in results 30-40 cm eels were used (de Boer and Hagel, 1993).
Almost all samples consisted of a pool of equal weights of fillets or organs of 25 fishes. Pooled samples were used to reduce the variation in results. Normally only the edible part of the fish was analysed, but sometimes livers or other organs were used for analysis. Unless the organs are mentioned, Table 1 shows PBDE concentrations in the edible part (fillet/muscle tissue) of the fish.
Sediment samples were taken by a Van Veen- sampler, according to the draft guideline NPR6600. Pooled samples, consisting of 10 sub-samples from an area of 100 m^2, were analysed. All sample locations, corresponding with the location numbers in Table 1, are shown in Fig. 1 and 2.

2.2. Methods

All samples were extracted according to the Soxhlet-method with n-pentane/dichloromethane (1:1). Samples analysed before 1983 were only extracted with n-pentane. Because for lean fish tissues, extraction efficiencies of pentane-extraction are less than those of pentane/dichloromethane extraction, PBDE-concentrations in lean tissues analysed before 1983 may be biased to the low side. For fatty tissues (> approximately 100 g/kg fat) no differences in results are expected (de Boer, 1988b).
After removal of dichloromethane on the rotary evaporator, the fat-extracts were cleaned-up on alumina columns and fractionated on silica columns (de Boer, 1988b). PBDEs were present in the second fraction of the silica column eluate together with most chlorinated pesticides.
The final analyses, identification and quantification, was carried out by capillary gas chromatography with electron capture detection (^{63} Ni). Stationary phases used in the fused silica capillary columns were non-polar (CP-Sil 8) or medium-polar (CP-Sil 19). Lengths and diameters were gradually optimized. Since 1988 50m columns with 0.15 mm internal diameters were used (de Boer and Dao, 1989).
Because of a lack of commercially available individual PBDE congeners, the technical mixture Bromkal 70 5DE was used as a standard. By gas chromatographic analysis of this mixture with flame ionisation detection, the composition of this mixture could be estimated. Recently we have received pure standards of 2,4,2',4'-tetrabromodiphenylether (TBDE) and 2,4,5,2'-,4'-pentabromodiphenylether (PeBDE) from Prof. Dr. A. Bergman of the Wallenberg Laboratory, University of Stockholm. Comparison of the Bromkal standard used with these Swedish standards showed that the percentage of TBDE in this mixture was 36.1% and of PeBDE 35,5%. A correction of the initially estimated data of - 5.6% for TBDE and -8.9% for PeBDE was therefore, necessary. The data given in this report have all been corrected on the basis of this comparison. In order to control the reproducibility of the method, from 1991 an internal laboratory reference material (cod liver) was analysed on TBDE simultaneously with each series of samples. The results were plotted in a quality control chart (Fig. 3). In these charts, twice the standard deviation of the first 10 results taken as a warning limit and three times the standard deviation is taken as an alarm-level. Certified reference materials for PBDEs are not available. Detection limits varied between <0.1 and <20 µg/kg, dependent of the dilution or concentration of the extract and the presence of interfering peaks.

3. Results and discussion

From the quality control chart in Fig. 3 a standard deviation of 20% is obtained for the analysis of TBDE since 1991. Because only minor modifications of the method have taken place, this standard deviation is representative for all TBDE results. Because mostly occurring in lower concentrations, the standard deviation for PeBDE may be higher. By the use of pooled samples, the effect of natural variation on the PBDE results is reduced. Nevertheless sampling will always be a source of variation. We have estimated this variation for PCBs in yellow eel, by analysing 25 individual eels from 4 locations (de Boer and Hagel, 1993). Standard deviations varied per location between 28 and 61%. The 95% confidence intervals for the results of pooled samples, related to these standard deviations by (rsd x 1.9)/√n, varied between 11 and 20%. The analytical error is included in this confidence interval. Because for TBDE and PeBDE similar methods as for PCBs are used in yellow eel. Given the analytical standard deviation of 20% for TBDE the 95%

confidence interval of the TBDE results in this report may be estimated at 25-30%. Given the expected larger analytical error for PeBDE, the 95% confidence interval for PeBDE results in yellow eel in this report is estimated at 30-40%.
For other samples than eel, no attention was paid to the selection of a specific length class and the sampling period. Therefore a larger natural variation may be expected in TBDE and PeBDE results. 95% Confidence intervals for TBDE and PeBDE in other samples are estimated at 30-40% for TBDE and 40-50% for PeBDE. Mainly because of its non-migrating behaviour, yellow eel is very suitable to serve as a bio-indicator in passive biological monitoring. Because a restricted length class was sampled each year, a biomaginification effect could be neglected. Therefore, temporal trend graphs could easily be constructed (Fig. 4 and 5).
For cod a biomaginification effect was found for certain compounds (higher chlorinated PCBs, p,p'-DDE), but not for PBDEs. Therefore, although different length classes of cod were sampled, no correction was needed for the TBDE values, prior to constructing temporal trends graphs for the different parts of the North Sea. These graphs are shown in Fig. 6.

Table 1 shows the concentrations of TBDE and PeBDE in all samples analysed on a wet weight basis, together with the fat contents of the biological samples. In Table 2 the TBDE concentrations in sediments are given together with the dry weights and total organic carbon contents of the samples.
Extremely high TBDE and PeBDE concentrations have been found in cormorant liver and kidney from the Biesbosch (upto 25,000 µg/kg TBDE and 4,000 µg/kg PBDE (wet weight)). This was only one measurement in a single cormorant. TBDE-concentrations in eel from the Hollands Diep around that time were close to 200 µg/kg wet weight. Translated to a lipid weight basis a biomaginification factor of 800 is found. Pieters (1991) reported biomaginification factors of upto 5000 for PCBs between cormorant livers and freshwater mussels from the Ketelmeer, although a broad variation in biomaginification factors was found. It would be interesting to see if more cormorants or other birds would also contain such high PBDE-concentrations.
Biomaginification is also observed in dolphins and porpoise from the southern North Sea and the Atlantic west of Ireland. Biomaginification factors between fish and these marine mammals are approximately 10-30.
All other TBDE-concentrations in aquatic biota are clearly lower than 1,000 µg/kg wet weight and most TBDE-concentrations even lower than 100 µg/kg. Most PeBDE-concentrations in aquatic biota are lower than 10 µg/kg.
Temporal trend graphs all shows overall decreasing trends for TBDE-concentrations during the last decade, except in eel from the river Roer, where a clear increasing trend is shown (Fig. 4-6). There may be a relation between this increase and a possible use of these compounds in hydraulic systems in German mining areas. Because the relatively low flux of the river Roer, the PBDE-input of this river in the river Meuse is very restricted. Nevertheless, the TBDE-trend in yellow eel from the Meuse at Keizersveer seems to increase slightly since 1989. PBDE-levels in the river Meuse varied strongly during the last decade. Peak concentrations are observed in 1986-1987. PBDE-concentrations in the river Rhine were clearly higher than in the river Meuse, showing that the main PBDE-load in the Netherlands freshwater systems is originating from Germany. PBDE-concentrations have decreased during the last decade, although Fig. 4 shows a stabilisation or even again a slight increase since 1989 in yellow eel from the locations Rhine, Lobith, Hollands Diep and Yssel Lake.

Temporal TBDE trends in cod liver from the North Sea are slightly decreasing. The number of data is, however, very limited.
In the sediment samples PeBDE-concentration are higher than the TBDE-concentrations. The PBDE-pattern in sediments is more comparable to the pattern of the technical mixture Bromkal 70-5DE. The higher TBDE-concentrations in fishes may be caused by a selective uptake of lower brominated compounds. Bruggeman et al. (1984) suggested that a membrane barrier would exist for higher brominated compounds due to the larger size of the molecules. Therefore, in sediments more higher brominated compounds may be expected. These compounds were not studied until now. A preliminary mass spectrometric analysis in river Rhine sediment showed the presence of hexabromodiphenylethers.
Dividing TBDE-concentrations expressed on a lipid weight basis in yellow eel, by TBDE-concentrations expressed on a total organic carbon basis in sediments from the same location (Table 2) result for all 4 locations in ratios of 0.5 - 0.7. For PCBs these ratios are normally between 1 and 4 (Bruggeman et al., 1989). This shows that, although only based on 4 locations, TBDE tends to adsorb better to the sediment, compared to PCBs.
TBDE-levels in eel from the river Rhine and Meuse are comparable with TBDE-concentrations in Arctic char from Lake Vättern in Sweden: 400 μg/kg lipid weight. Some pike samples from Swedish rivers contained extremely high TBDE-concentrations: upto 24 mg/kg lipid weight (Janssen et al., 1993). TBDE-concentrations in herring from the Skagerrak were comparable to TBDE-concentrations in North Sea fish. Baltic seals contained higher TBDE-concentrations then North Sea seals (Jenssen et al., 1987).

Conclusions

A large database on concentrations of 2,4,2',4'-TBDE and 2,4,5,2',4'-PeBDE in aquatic biota and sediments is presented. 95% confidence intervals of the data, almost all determined in pooled samples, are estimated at 25-40% for TBDE and PeBDE in yellow eel and 30-50% for TBDE and PeBDE in all other samples.
Extremely high PBDE levels were found in organs of a cormorant from the Biesbosch in 1981. These data suggest a high biomaginification of PBDEs in these birds. This should be confirmed by additional data. Also in marine mammals biomaginification is found, but on a lower scale than for the cormorant. Most TBDE-concentrations in fish and shellfish are lower than 100 μg/kg, most PeBDE-concentrations are lower than 10 μg/kg.
Higher PBDE-concentrations in the river Rhine show that the main PBDE-load in Dutch freshwater ecosystems in originating from Germany. Most temporal trend graphs show decreasing trends, although a stabilisation is visible in eel from the river Rhine and Meuse since 1988/1989. An increasing TBDE-trend is found in yellow eel from the river Roer. This could be related to the use of PBDE in hydraulic mining equipment in Germany. Higher TBDE-concentrations in biological samples, in comparison with sediments, are presumably caused by a selective uptake of TBDE due to a membrane barrier for higher brominated compounds. PBDEs seem to show a better absorbtion to the sediment than PCBs. More data on PBDEs in sediments are needed to confirm this result. Also, it may be worthwhile to investigate the presence of higher brominated PBDEs in sediments.

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Table 1: Concentrations of 2,4,2',4'-TBDE and 2,4,5,2',4'-PeBDE, expressed in µg/kg wet weight.

No.	Location	Year	Species	TBDE (µg/kg)	PeBDE (µg/kg)	Lipid (g/kg)
1	Aar Kanaal (Ter Aar)	1992	Yellow eel	6.2	<1	152
2	Amstel Drecht Kanaal	1991	Yellow eel	<1	0.5	124
3	Amsterdam-Rijnkanaal (Diemen)	1992	Yellow eel	3.5		66
4	Apeldoorns Kanaal (Hattem)	1991	Yellow eel	5	1.3	274
5	Atlantic, south of Ireland	1987	Hake	0.8	0.4	640
6	Atlantic, west of Ireland	1983	Dolphin blubber	590	<10	836
6	Atlantic, west of Ireland	1983	Dolphin muscle	18		31
6	Atlantic, west of Ireland	1986	Hake liver	<20	<10	539
7	Bay of Biscay	1983	Hake	69		448
7	Bay of Biscay	1983	Hake liver	70		373
8	Bergsche plas	1991	Yellow eel	1.6	1	142
9	Biesbosch	1981	Cormorant kidney	18,000	2,000	22
9	Biesbosch	1981	Cormorant liver	25,000	4,000	31
10	Binnen Liede	1983	Yellow eel	<10	<10	174
11	Boven Merwede (Gorinchem)	1990	Roach	2.8		13
11	Boven Merwede (Gorinchem)	1989	Yellow eel	9.7	1.8	83
11	Boven Merwede (Gorinchem)	1990	Yellow eel	48		220
11	Boven Merwede (Gorinchem)	1991	Yellow eel	120	11	279
12	Buiten Liede	1983	Yellow eel	<10	<10	168
13	Callandkanaal (Europoort)	1985	Yellow eel	9.7	<10	102
14	Central North Sea	1985	Cod	0.2	<10	7.7
14	Central North Sea	1991	Cod	1	<0.1	9
14	Central North Sea	1983	Cod liver	42	5.8	487
14	Central North Sea	1984	Cod liver	73		569
14	Central North Sea	1987	Cod liver	53	3.9	569
14	Central North Sea	1988	Cod liver	58	13	502
14	Central North Sea	1989	Cod liver	12		220
14	Central North Sea	1985	Herring	1	<10	131
15	Danish west-coast (Esbjerg)	1989	Plaice	<0.1		7.4
15	Danish west-coast (Esbjerg)	1989	Plaice liver	1.1		85
16	Delfzijl (Buitenhaven)	1984	Yellow eel	3.5		133
17	Delfzijl (Zeehavenkan)	1984	Yellow eel	<10		125
18	Diemerzeedijk	1985	Yellow eel	<10	<10	133
18	Diemerzeedijk	1985	Yellow eel	<10	<10	158
19	Diemerzeedijk-Pampus	1985	Yellow eel	<10	<10	169
20	Eastern Scheldt	1984	Mussel	0.8	<10	18
20	Eastern Scheldt	1989	Mussel	0.4		21
20	Eastern Scheldt	1989	Mussel	0.3		15
20	Eastern Scheldt	1991	Mussel	0.3	<1	17
20	Eastern Scheldt	1991	Mussel	0.7		24
20	Eastern Scheldt	1991	Oyster	0.7	0.7	10
20	Eastern Scheldt	1984	Shrimp	0.3	<10	12
21	Egmond	1984	Shrimp	0.7	<10	15
21	Egmond	1984	Shrimp	1.5	<10	23
22	English Channel	1982	Hake liver	11	<10	615
22	English Channel	1982	Sprat	1.8		123
23	English Channel(west)	1989	Plaice	0.4		8.7
23	English Channel (west)	1989	Plaice liver	4.5		110
24	English east-coast	1989	Plaice	<0.1		7.4
24	English east-coast	1989	Plaice liver	6.6		160
25	German Bight	1991	Dab	0.19	<0.1	16
25	German Bight	1991	Dab liver	3	<2	356
25	German Bight	1989	Plaice	0.1		7.3
25	German Bight	1989	Plaice liver	2.1		180
25	German Bight	1990	Sole	<0.1	<0.1	17
25	German Bight	1990	Sole liver	2	<2	125
26	Geul (Meersen)	1992	Yellow eel	6.8	0.7	114
27	Haringvliet-east	1977	Yellow eel	97	<10	155
27	Haringvliet-east	1978	Yellow eel	88	<10	162
27	Haringvliet-east	1979	Yellow eel	180	<10	154
27	Haringvliet-east	1984	Yellow eel	190		173
27	Haringvliet-east	1985	Yellow eel	48	5	223
27	Haringvliet-east	1986	Yellow eel	34	6.5	261
27	Haringvliet-east	1987	Yellow eel	85	<10	217
27	Haringvliet-east	1988	Yellow eel	88	<2	163

27	Haringvliet-east	1990	Roach	16		21
27	Haringvliet-east	1992	Sediment	6.7	7.3	
27	Haringvliet-east	1988	Yellow eel	70	<4	211
27	Haringvliet-east	1989	Yellow eel	62		210
27	Haringvliet-east	1990	Yellow eel	33		164
27	Haringvliet-east	1991	Yellow eel	32	1.1	76
27	Haringvliet-east	1992	Yellow eel	40	3.4	118
28	Haringvliet-east (Bridge)	1991	Yellow eel	45	3.6	119
29	Haringvliet-east (Hitsersekade)	1991	Yellow eel	74	4.3	148
30	Haringvliet-east (Nieuwe Dijk)	1991	Yellow eel	77	<2	134
31	Haringvliet-west (Hellevoetsluis)	1989	Yellow eel	41	<2	149
31	Haringvliet-west (Hellevoetsluis)	1990	Yellow eel	32		170
31	Haringvliet-west (Hellevoetsluis)	1991	Yellow eel	62		141
31	Haringvliet-west (Hellevoetsluis)	1991	Yellow eel	48	2.1	189
31	Haringvliet-west (Hellevoetsluis)	1992	Yellow eel	22	<2	113
32	Haringvliet-west (Middelharnis)	1991	Yellow eel	40	1.7	90
33	Hollands Diep	1991	Pike-perch	5.5		8.6
33	Hollands Diep	1990	Pike-perch liver	61	19	67
33	Hollands Diep	1990	Pike-perch	5.1	1.3	12
33	Hollands Diep	1979	Yellow eel	190	<10	233
33	Hollands Diep	1983	Yellow eel	130	<10	186
33	Hollands Diep	1984	Yellow eel	150		149
33	Hollands Diep	1985	Yellow eel	47	3.7	172
33	Hollands Diep	1986	Yellow eel	59	2.7	235
33	Hollands Diep	1987	Yellow eel	110	<10	239
33	Hollands Diep	1988	Yellow eel	77	4	196
33	Hollands Diep	1989	Yellow eel	32	2.6	183
33	Hollands Diep	1990	Yellow eel	43		170
33	Hollands Diep	1991	Yellow eel	66	2.8	155
33	Hollands Diep	1991	Yellow eel	110		320
33	Hollands Diep	1991	Yellow eel	63	2.8	129
33	Hollands Diep	1992	Yellow eel	60	1	151
34	Hollandse IJssel (Gouderak)	1990	Pike-perch	25	4.7	51
34	Hollandse IJssel (Gouderak)	1990	Pike-perch	5.6	1	8.9
34	Hollandse IJssel (Gouderak)	1990	Pike-perch liver	25	4.7	51
34	Hollandse IJssel (Gouderak)	1984	Yellow eel	91		201
34	Hollandse IJssel (Gouderak)	1987	Yellow eel	52	<10	130
35	IJ, Amsterdam	1992	Yellow eel	4.3		123
36	IJmond	1991	Shrimp	0.1		27
37	Irish Sea	1982	Hake liver	18	<10	551
38	Ketelmeer	1991	Yellow eel	16	<2	137
38	Ketelmeer	1990	Roach	1.8		14
38	Ketelmeer	1987	Silver eel	7.4	4.9	270
38	Ketelmeer	1987	Silver eel	15	4.3	350
38	Ketelmeer	1987	Silver eel	81	14	370
38	Ketelmeer	1987	Silver eel	39	6.5	310
38	Ketelmeer	1977	Yellow eel	79	<10	250
38	Ketelmeer	1978	Yellow eel	54	<10	242
38	Ketelmeer	1980	Yellow eel	110	<10	243
38	Ketelmeer	1981	Yellow eel	88	<10	239
38	Ketelmeer	1982	Yellow eel	120	<10	311
38	Ketelmeer	1984	Yellow eel	100		276
38	Ketelmeer	1985	Yellow eel	70	7.9	276
38	Ketelmeer	1987	Yellow eel	72	<4	291
38	Ketelmeer	1988	Yellow eel	54	4.1	279
38	Ketelmeer	1989	Yellow eel	17	2	195
38	Ketelmeer	1990	Yellow eel	21		234
38	Ketelmeer	1992	Yellow eel	33	2.3	192
39	Lauwersmeer	1988	Yellow eel	3.4	2.2	190
39	Lauwersmeer	1992	Yellow eel	1.7	<1	138
40	Lek (Culemborg)	1991	Yellow eel	34	3.5	123
40	Lek (Culemborg)	1992	Yellow eel	70	3.8	157
41	Lek (Krimpen)	1988	Yellow eel	76	2.4	235
41	Lek (Krimpen)	1989	Yellow eel	53		222
42	Lek (Lekkerkerk)	1990	Yellow eel	97	<4	229
43	Linge (Rhenoij)	1991	Yellow eel	12	0.6	143
44	Maas-Waalkanaal (Malden)	1992	Yellow eel	40	2.2	262
45	Maasvlakte	1984	Shrimp	1	<10	14

46	Markermeer	1991	Yellow eel	4	<1	153
46	Markermeer	1992	Yellow eel	6.2	<1	206
47	Meuse (Borgharen)	1992	Yellow eel	11	1.4	248
48	Meuse (Eijsden)	1983	Yellow eel	72	<10	120
48	Meuse (Eijsden)	1984	Yellow eel	71		161
48	Meuse (Eijsden)	1985	Yellow eel	<10	<10	120
48	Meuse (Eijsden)	1986	Yellow eel	53		143
48	Meuse (Eijsden)	1987	Yellow eel	35	<4	101
48	Meuse (Eijsden)	1988	Yellow eel	<4	<4	145
48	Meuse (Eijsden)	1990	Yellow eel	1.3	<2	102
48	Meuse (Eijsden)	1991	Yellow eel	5.1	<2	150
48	Meuse (Eijsden)	1992	Yellow eel	3.6	<1	140
49	Meuse (Keizersveer)	1984	Yellow eel	80		244
49	Meuse (Keizersveer)	1985	Yellow eel	<10	<10	219
49	Meuse (Keizersveer)	1986	Yellow eel	6.7	<1	96
49	Meuse (Keizersveer)	1987	Yellow eel	88	<10	236
49	Meuse (Keizersveer)	1988	Yellow eel	25		191
49	Meuse (Keizersveer)	1989	Yellow eel	13	<2	216
49	Meuse (Keizersveer)	1992	Sediment	6.9	8.2	
49	Meuse (Keizersveer)	1989	Yellow eel	8.8	1.2	111
49	Meuse (Keizersveer)	1990	Yellow eel	27	<4	201
49	Meuse (Keizersveer)	1991	Yellow eel	15	<1	142
49	Meuse (Keizersveer)	1992	Yellow eel	25	<1	170
50	Meuse (Lith)	1989	Sea trout	1.8	0.2	31
50	Meuse (Lith)	1989	Sea trout	2.1	0.6	41
51	Meuse (Venlo)	1989	Yellow eel	18	2.8	273
52	Meuse (Urmond)	1986	Yellow eel	110		230
53	Niers	1984	Yellow eel	<10		227
54	Nieuwe Maas (Brienenoord)	1989	Yellow eel	55	4.3	228
55	Nieuwe Maas (Schiedam)	1989	Yellow eel	18	1.1	170
56	Nieuwe Merwede	1992	Sediment	17		
56	Nieuwe Merwede	1987	Yellow eel	97	<4	207
56	Nieuwe Merwede	1988	Yellow eel	77	2.4	194
56	Nieuwe Merwede	1989	Yellow eel	40	3.8	205
56	Nieuwe Merwede	1991	Yellow eel	84	4.4	180
56	Nieuwe Merwede	1992	Yellow eel	85	8.7	172
56	Nieuwe Merwede	1989	Yellow eel liver	5.7	0.61	40
57	Nieuwe Waterweg (Vlaardingen)	1991	Yellow eel	25	1.3	150
58	Noordhollands kanaal (Akersloot)	1992	Yellow eel	2.4		93
59	Noordzeekanaal (Hembrug)	1984	Yellow eel	<10		111
59	Noordzeekanaal (Hembrug)	1992	Yellow eel	4.3	<0.5	94
60	Noordzeekanaal (Kruithaven)	1992	Yellow eel	5.2		73
61	Noordzeekanaal (Velsen)	1992	Yellow eel	3.3	1.1	113
62	Northern North Sea	1986	Cod	0.4	<10	14.9
62	Northern North Sea	1983	Cod liver	30	5.1	499
62	Northern North Sea	1986	Cod liver	17	<10	492
62	Northern North Sea	1987	Cod liver	23	1.5	660
62	Northern North Sea	1989	Cod liver	14	1.3	650
62	Northern North Sea	1985	Herring	0.7	<10	26
63	North Sea (IJmuiden)	1990	Dab	3.5	<0.3	20
64	Oostvaardersplassen	1984	Yellow eel	<10	<10	248
65	Oude Rijn Sprangen	1986	Yellow eel	3.9	<4	196
66	Oude Maas (Rhoon)	1989	Yellow eel	77		227
66	Oude Maas (Rhoon)	1990	Yellow eel	110	<5	164
68	Paterswoldermeer	1991	Yellow eel	1.9	<4	291
69	Prinses Margrietkanaal (Suawoude)	1992	Yellow eel	1.1	<1	178
70	Rhine (Lobith)	1990	Roach	2.4		14
70	Rhine (Lobith)	1984	Yellow eel	250		199
70	Rhine (Lobith)	1985	Yellow eel	71	7.5	137
70	Rhine (Lobith)	1986	Yellow eel	63	5.9	201
70	Rhine (Lobith)	1987	Yellow eel	97	<4	143
70	Rhine (Lobith)	1988	Yellow eel	70	7.4	176
70	Rhine (Lobith)	1989	Yellow eel	18	1.9	112
70	Rhine (Lobith)	1990	Yellow eel	29		135
70	Rhine (Lobith)	1991	Yellow eel	18	0.9	63
70	Rhine (Lobith)	1992	Yellow eel	39	2.1	110
71	Rijnmond	1984	Shrimp	2.5	<10	15
72	Ringvaart (Haarlemmermeer)	1983	Yellow eel	<10	<10	165

73	Roer (Vlodrop)	1983	Yellow eel	110	<10	254
73	Roer (Vlodrop)	1984	Yellow eel	120		222
73	Roer (Vlodrop)	1986	Yellow eel	68	5.4	124
73	Roer (Vlodrop)	1987	Yellow eel	190		301
73	Roer (Vlodrop)	1988	Yellow eel	260	32	283
73	Roer (Vlodrop)	1989	Yellow eel	180	15	148
73	Roer (Vlodrop)	1990	Yellow eel	130	<4	186
73	Roer (Vlodrop)	1991	Yellow eel	220	11	163
73	Roer (Vlodrop)	1992	Yellow eel	240	21	192
74	Rottige Meenthe	1988	Yellow eel	1.1	<1	192
75	Shetland Islands	1991	Mackerel	3.1	<1	140
76	Skagerrak	1989	Plaice	0.1		7.1
76	Skagerrak	1989	Plaice liver	1.3		98
76	Skagerrak	1991	Herring	4.3	1.7	220
77	Southern North Sea	1992	Blenny	1	0.2	22
77	Southern North Sea	1992	Brill	0.4	<0.1	11
77	Southern North Sea	1992	Brill liver	13	0.7	236
77	Southern North Sea	1984	Cod	0.4	<10	5.6
77	Southern North Sea	1991	Cod	0.5	<0.1	
77	Southern North Sea	1991	Cod	1		5.9
77	Southern North Sea	1992	Cod	0.3	<0.1	7.9
77	Southern North Sea	1981	Cod liver	460		465
77	Southern North Sea	1982	Cod liver	180	17	539
77	Southern North Sea	1984	Cod liver	140		443
77	Southern North Sea	1984	Cod liver	130	<10	538
77	Southern North Sea	1987	Cod liver	130	2.8	491
77	Southern North Sea	1989	Cod liver	45	1.7	610
77	Southern North Sea	1991	Cod liver	110	3	483
77	Southern North Sea	1990	Dolphin blubber	3000	220	696
77	Southern North Sea	1990	Dolphin blubber	2600		817
77	Southern North Sea	1990	Dolphin kidney	44	7.9	29
77	Southern North Sea	1990	Dolphin liver	45	5.3	43
77	Southern North Sea	1990	Dolphin liver	180	30	53
77	Southern North Sea	1990	Dolphin muscle	57	12	20
77	Southern North Sea	1990	Dolphin spleen	43	8.7	28
77	Southern North Sea	1985	Herring	1.6	<10	45
77	Southern North Sea	1985	Herring	1.7	<10	11
77	Southern North Sea	1991	Herring	11		191
77	Southern North Sea	1990	Porpoise blubber	830	79	806
77	Southern North Sea	1989	Shrimp	0.4	0.1	21
77	Southern North Sea	1992	Shrimp	<0.1	<0.1	17
77	Southern North Sea	1985	Shrimp liver	4	<4	62
77	Southern North Sea	1992	Smelt	1.2	0.2	18
77	Southern North Sea	1991	Sole	0.5		21
77	Southern North Sea	1992	Sole	0.1	<0.1	10
77	Southern North Sea	1992	Turbot	0.2	<0.1	13
77	Southern North Sea	1992	Turbot liver	7	1	95
77	Southern North Sea	1987	Twaite shad	77	<4	28.1
77	Southern North Sea	1991	Twaite shad liver	15	1.7	159
77	Southern North Sea	1992	Whiting	0.4	0.1	9.5
78	Southern North Sea (Vlaamse Bank)	1992	Herring	28	17	107
78	Southern North Sea (Vlaamse Bank)	1992	Herring liver	2.4	1.3	25
79	Straits of Dover	1985	Herring	0.9	<10	50
79	Straits of Dover	1985	Herring	7.6	<10	126
79	Straits of Dover	1989	Plaice	0.2		8.7
80	Tjeukemeer	1988	Yellow eel	<2		167
80	Tjeukemeer	1991	Yellow eel	5.3	<2	114
81	Tongelreep (Bruggerhuizen)	1992	Yellow eel	7.6	<2	224
82	Twentekanaal (Almelo)	1987	Roach	5.7	<1	12
82	Twentekanaal (Almelo)	1987	Yellow eel	20	<4	192
83	Twentekanaal (Hengelo)	1987	Roach	15	<1	22
83	Twentekanaal (Hengelo)	1987	Yellow eel	<5	<10	169
83	Twentekanaal (Hengelo)	1991	Yellow eel	9.7	2.9	165
83	Twentekanaal (Hengelo)	1992	Yellow eel	4.7	<1	142
84	Twentekanaal (Lochem)	1987	Yellow eel	49	<2	138
85	Utrecht	1983	Human milk	0.4		426
86	Vecht (Ommen)	1991	Yellow eel	7.7	<1	163
86	Vecht (Ommen)	1992	Yellow eel	6.6	0.5	173

87	Vliet (Rijswijk)	1988	Yellow eel	<3	<5	157
88	Volkerak	1986	Yellow eel	4.9		109
88	Volkerak	1987	Yellow eel	13	<1	97
88	Volkerak	1988	Yellow eel	7.7	<4	115
88	Volkerak	1989	Yellow eel	14	<4	105
88	Volkerak	1992	Yellow eel	14	3.4	146
89	Waal (Heesselt)	1989	Sea trout	3.3	0.5	36
89	Waal (Heesselt)	1989	Sea trout	2.9	0.7	37
90	Waal (Nijmegen)	1983	Yellow eel	340	<10	250
90	Waal (Nijmegen)	1984	Yellow eel	160		233
90	Waal (Nijmegen)	1985	Yellow eel	46	7.5	195
91	Waal (Tiel)	1990	Roach	2.1		11
91	Waal (Tiel)	1992	Sediment	23	21	
91	Waal (Tiel)	1987	Silver eel	55	4.4	329
91	Waal (Tiel)	1983	Yellow eel	180	<10	215
91	Waal (Tiel)	1984	Yellow eel	330		222
91	Waal (Tiel)	1985	Yellow eel	110	15	238
91	Waal (Tiel)	1986	Yellow eel	55	6.1	111
91	Waal (Tiel)	1987	Yellow eel	170	<10	282
91	Waal (Tiel)	1988	Yellow eel	110	10	243
91	Waal (Tiel)	1989	Yellow eel	79		251
91	Waal (Tiel)	1990	Yellow eel	43		240
91	Waal (Tiel)	1991	Yellow eel	130	22	261
91	Waal (Tiel)	1992	Yellow eel	110	6.3	209
92	Wadden Sea-east (Eems)	1984	Mussel	0.4	<10	12
92	Wadden Sea-east (Eems)	1984	Shrimp	<10	<10	16
92	Wadden Sea-east (Eems)	1992	Yellow eel	1.5	1.5	116
93	Wadden Sea (Steendiep)	1991	Dab	0.4	<0.1	20
93	Wadden Sea (Steendiep)	1991	Dab liver	11	<1	364
93	Wadden Sea (Steendiep)	1984	Mussel	0.4	<10	10
93	Wadden Sea (Steendiep)	1984	Shrimp	0.6	<10	14
93	Wadden Sea (Steendiep)	1991	Yellow eel	5.5	0.68	132
93	Wadden Sea (Steendiep)	1992	Yellow eel	9.7	<1	156
94	Western Scheldt (Hansweert)	1992	Yellow eel	6.3	0.8	114
95	Western Scheldt (Terneuzen)	1983	Yellow eel	3.5	<10	58
96	Western Scheldt (Hoedekenskerke)	1984	Shrimp	1	<10	10
97	Western Scheldt (Vlissingen)	1984	Mussel	1.5	<10	12
98	Yssel (Deventer)	1988	Yellow eel	40		176
98	Yssel (Deventer)	1989	Yellow eel	33	4	228
98	Yssel (Deventer)	1991	Yellow eel	56		220
98	Yssel (Deventer)	1992	Yellow eel	84	5.4	204
98	Yssel (Deventer)	1990	Yellow eel	110	<3	207
99	Yssel Lake (Medemblik)	1991	Pike-perch	1.1		12
99	Yssel Lake (Medemblik)	1984	Yellow eel	40		219
99	Yssel Lake (Medemblik)	1986	Yellow eel	15	<10	278
99	Yssel Lake (Medemblik)	1987	Yellow eel	22	<4	295
99	Yssel Lake (Medemblik)	1989	Yellow eel	12	1.4	342
99	Yssel Lake (Medemblik)	1991	Yellow eel	16	<2	311
99	Yssel Lake (Medemblik)	1991	Yellow eel	15		216
99	Yssel Lake (Medemblik)	1992	Yellow eel	22	<1	267
100	Yssel Lake (Afsluitdijk)	1990	Yellow eel	4.8		308
101	Yssel Lake (Ketelbrug)	1989	Yellow eel	22	2.1	283
102	Yssel Lake (Urk)	1990	Yellow eel	12		268
103	Zoommeer	1987	Yellow eel	3.8	<4	130
103	Zoommeer	1988	Yellow eel	<4		219
103	Zoommeer	1991	Yellow eel	3.1	<2	124
103	Zoommeer	1992	Yellow eel	3.4	<1	83
104	Zuid-Willemsvaart (Veghel)	1989	Yellow eel	3.7	1.5	186
105	Zuid-Willemsvaart (Weert)	1992	Yellow eel	3	0.6	84
106	Zuidlaardermeer (Noordlaren)	1992	Yellow eel	1.5	1.3	214

Table 2 - Concentrations of 2,4,2',4'-TBDE and 2,4,5,2',4'-PeBDE in sediments

Location	µg/kg wet weight		µg/kg dry weight		µg/kg org. C	
	TBDE	PeBDE	TBDE	PeBDE	TBDE	PeBDE
Haringvliet-east	6.7	7.3	11	12	410	450
Meuse, Keizerveer	6.9	8.2	11	13	250	300
Nieuwe Merwede	17	na	27	na	900	na
Waal, Tiel	23	21	36	33	990	910

na: not analysed

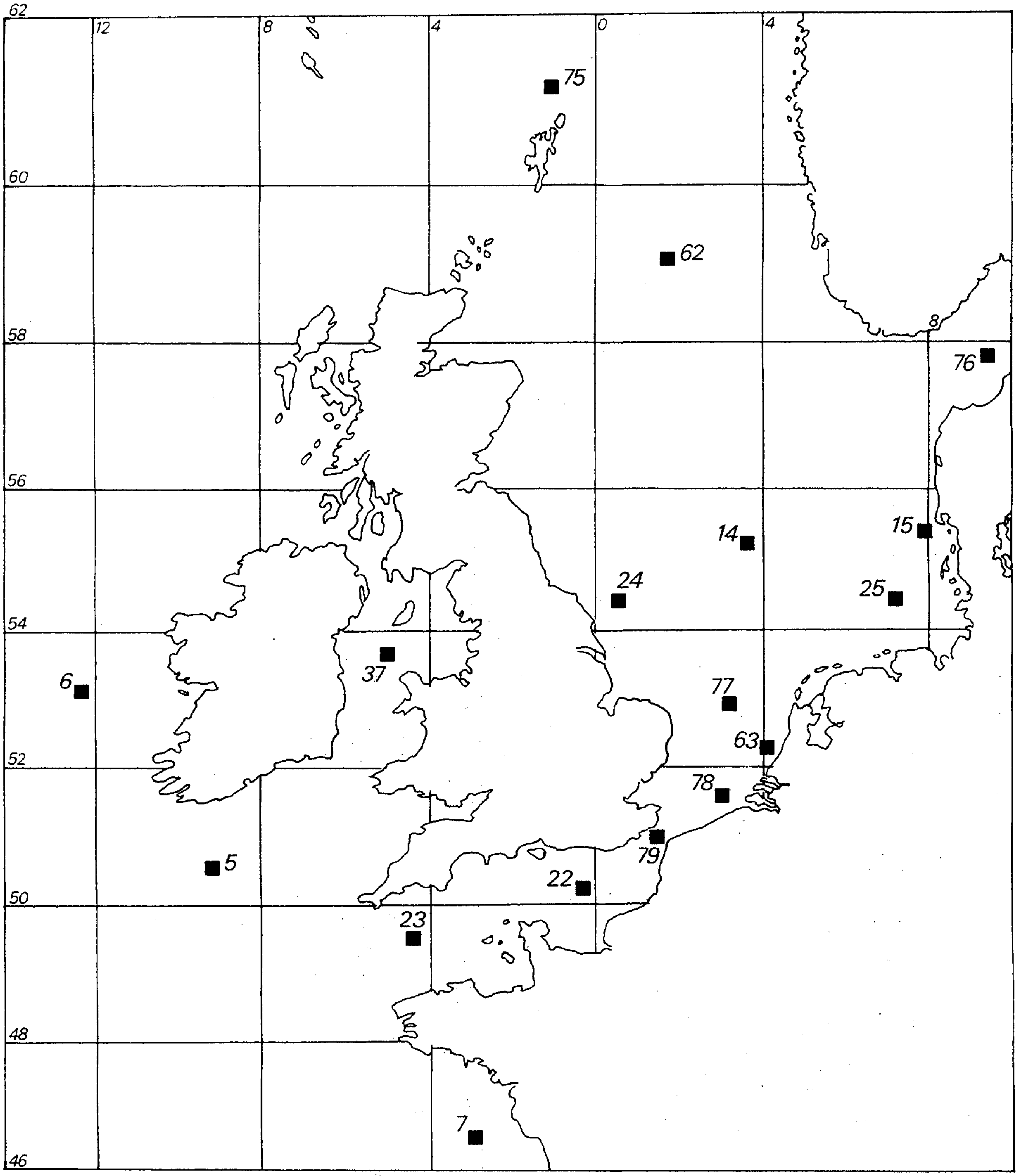


Figure 1 - Sample locations in the Netherlands.

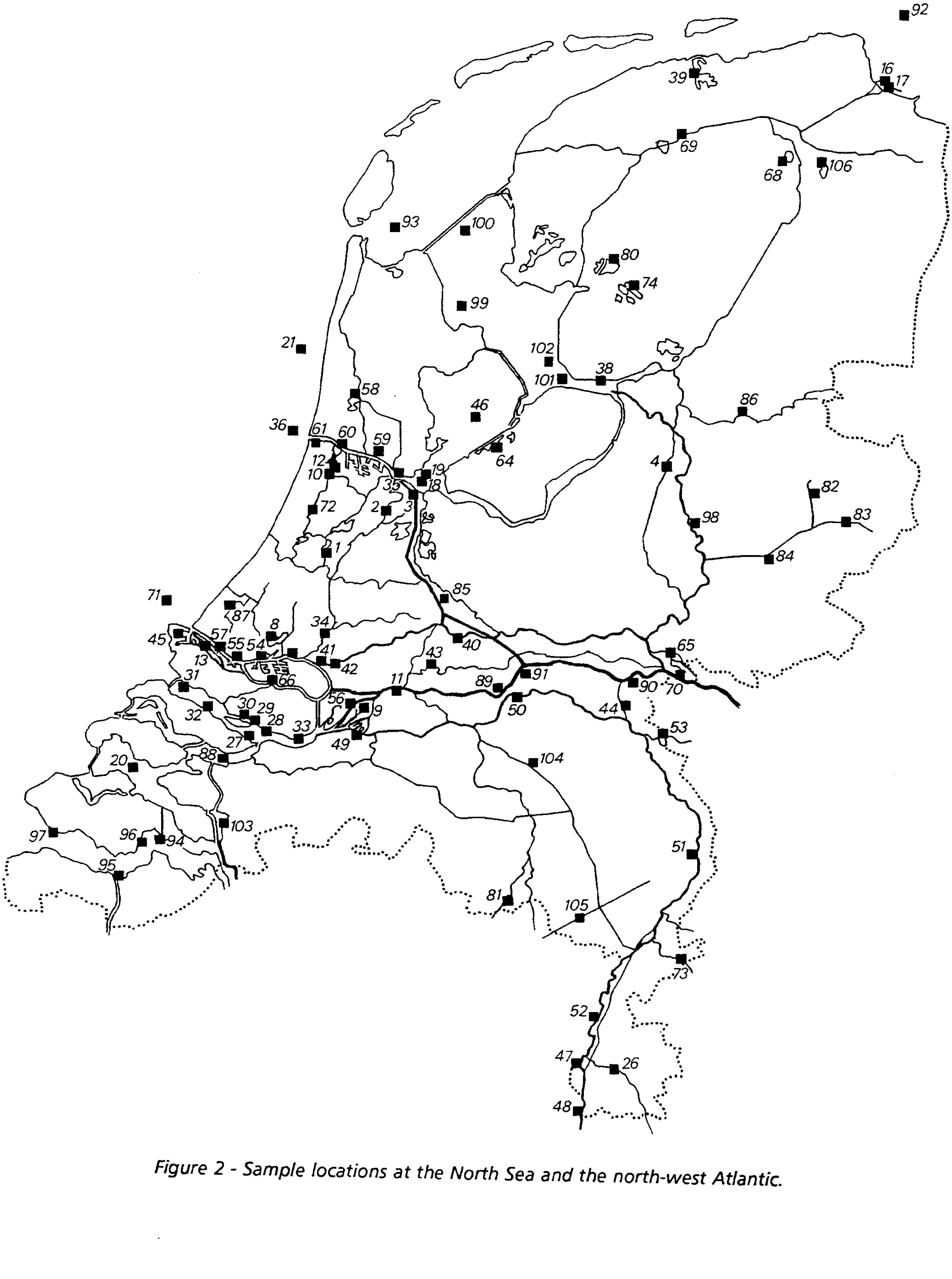


Figure 2 - Sample locations at the North Sea and the north-west Atlantic.

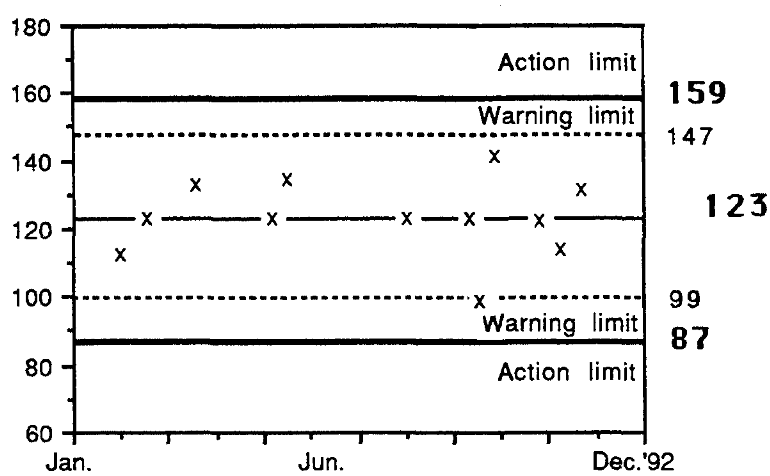


Figure 3 - Quality control chart of TBDE.

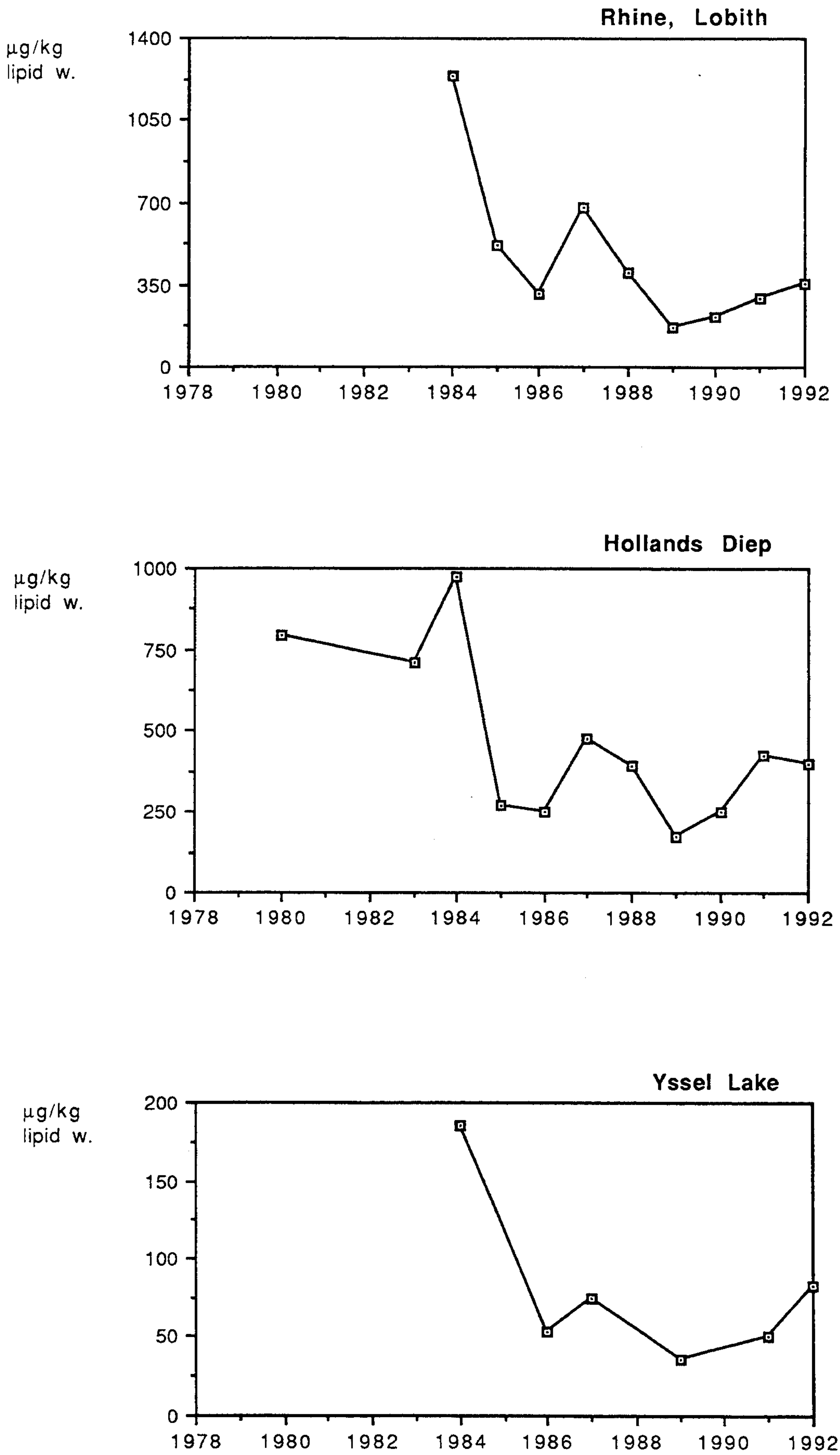
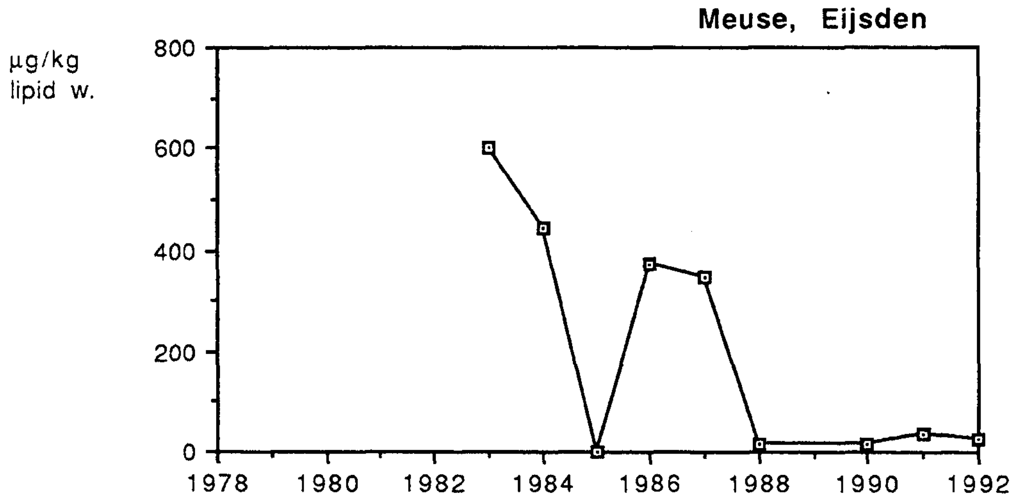
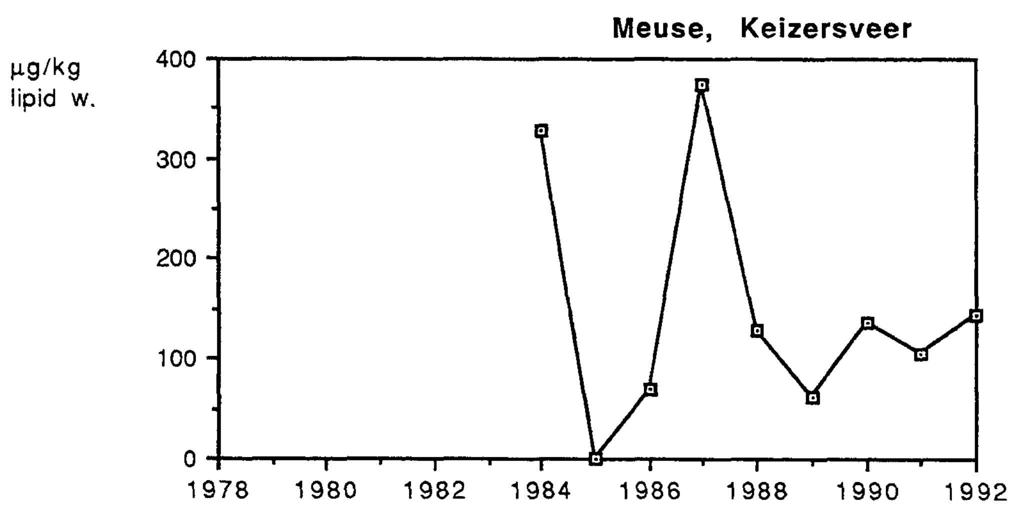


Figure 4 - Temporal trends of TBDE in yellow eel from the river Rhine (Lobith), the Hollands Diep and the Yssel Lake (Medemblik).





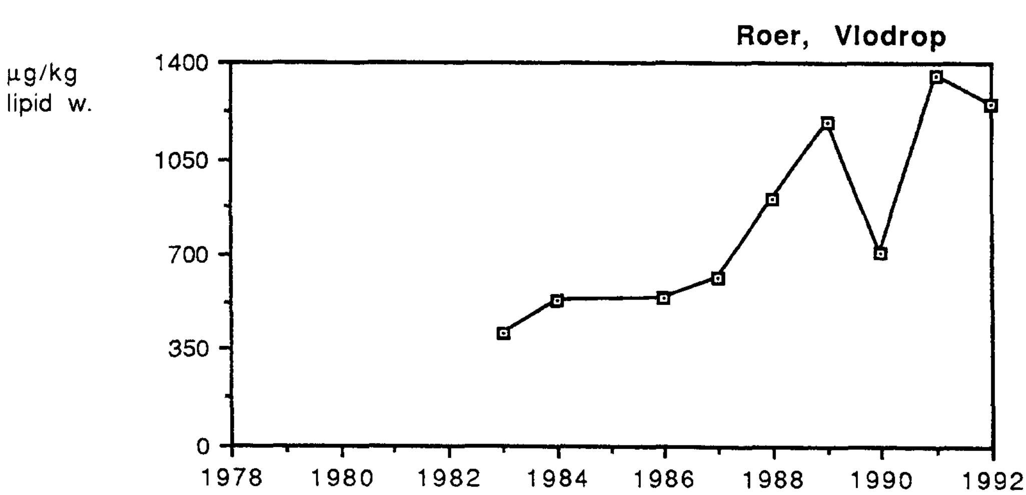


Figure 5 - Temporal trends of TBDE in yellow eel from the river Meuse (Eijsden), the river Meuse (Keizersveer) and the river Roer (Vlodrop).

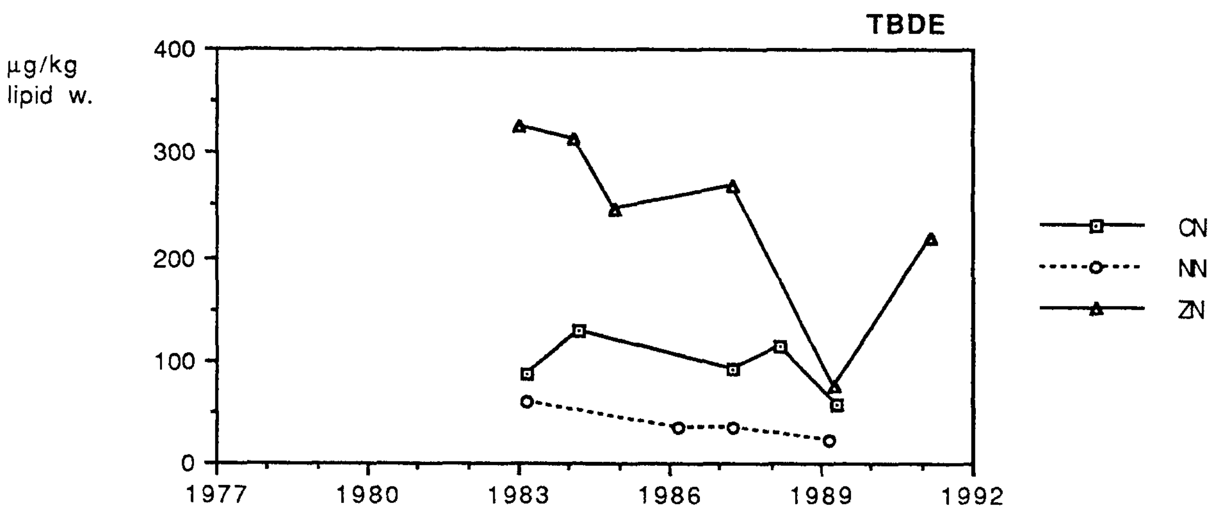


Figure 6 - Temporal trend of TBDE in cod liver from the northern (NN), central (CN) and southern North Sea (ZN).